

Anomalous double peak structure in Nb/Ni superconductor/ferromagnet tunneling DOS

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We have experimentally investigated the density of states (DOS) in Nb/Ni (S/F) bilayers as a function of Ni thickness, d_F . Our thinnest samples show the usual DOS peak at $\pm\Delta_0$, whereas intermediate-thickness samples have an anomalous “double-peak” structure. For thicker samples ($d_F \geq 3.5$ nm), we see an “inverted” DOS which has previously only been reported in superconductor/weak-ferromagnet structures. We analyze the data using the self-consistent non-linear Usadel equation and find that we are able to quantitatively fit the features at $\pm\Delta_0$ if we include a large amount of spin-orbit scattering in the model. Interestingly, we are unable to reproduce the sub-gap structure through the addition of any parameter(s). Therefore, the observed anomalous sub-gap structure represents new physics beyond that contained in the present Usadel theory.

The co-existence of superconductivity and ferromagnetism was first proposed by Fulde and Ferrell [1] and Larkin and Ovchinnikov [2] more than forty years ago. While some unusual materials have since been found with both superconducting and ferromagnetic transitions (*e.g.* ErRh₄B₄ [3]), much recent interest has focused on conventional superconductor/ferromagnet (S/F) proximity effect multi-layer systems. A wide variety of unusual phenomena has been proposed for these systems including oscillating critical temperatures, T_c [4], π -state Josephson junctions [5], and long-ranged odd-frequency triplet superconductivity [6].

Qualitative evidence for the first two of these effects is convincing, but definitive quantitative agreement with theory has been problematic. The evidence for triplet superconductivity is less certain, although a recent report by Keizer *et al* [7] provides tantalizing evidence for such an effect. One reason for the difficulty in achieving quantitative agreement with theory is the proliferation of physical effects that now have been incorporated into the theory, leading to a concomitant proliferation of fitting parameters, which makes discriminating fits to limited data sets nearly impossible.

In order to obtain more discriminating data sets and to further explore the SF proximity effect in the case of strong ferromagnets, we have undertaken superconducting tunneling densities of state (DOS) measurements on Nb/Ni thin-film bilayers. By varying the Ni thickness, d_F , we can track the spatial evolution of the behavior of the Cooper pairs diffusing into the ferromagnet. This approach gives us much more information per sample (the entire DOS spectrum) than T_c or J_c measurements, and is less sensitive to variations in boundary parameters. Analyzing these results with the most complete forms of the Usadel theory available has allowed us to discriminate

critically for the first time the relative importance of the various physical effects now incorporated into the theory. We find that by far the most important parameter beyond the exchange field, E_{ex} , is the degree of spin-orbit scattering (first suggested by Demler *et al* [8]). In addition, we find an anomalous double-peak structure in the DOS that has not been reported previously and that we have been unable to account for theoretically.

We use planar tunnel junctions of the form *normal-insulator-ferromagnet-superconductor*. A schematic of our sample geometry is shown in the inset of Fig. 2. The deposition of our samples and characterization of the tunnel junctions has been documented elsewhere [9, 10]. In brief, the various layers are sputtered and patterned with stencil masks *in situ* in a DC magnetron sputtering chamber without breaking vacuum. The Al₂O₃ tunnel barriers are formed by oxidizing the Al underlayer, and are canonical in their behavior, except for the zero-bias anomalies commonly observed in tunnel junctions incorporating magnetic materials; we discuss these below. We use a Co₆₀Fe₄₀ backing of the Al electrode so as to reduce its critical temperature below our lowest measurement temperature, which ensures normal/superconductor tunneling. We vary d_F from 0 nm to 5 nm in 0.5 nm increments. Ni has a Curie Temperature of roughly 600 K, and should be ferromagnetic for film thicknesses greater than two atomic layers (5 Å) [11, 12]. We have taken care to ensure that our Ni films are ferromagnetic by measuring the superconducting resistive transition of the bilayer in a parallel magnetic field: we detect hysteretic signals confirming the presence of magnetism in all our films with $d_F \geq 1.0$ nm.

Tunneling measurements were performed at 0.28 K using standard lock-in techniques. Samples were measured in zero magnetic field and also in a perpendicular field

greater than H_{c2} . In all of our samples, the normal-state conductance is a V-shaped curve in which the zero-bias conductance (ZBC) is roughly 1% – 2% lower than the conductance at 5 mV, which is typical of magnetic tunnel junctions [13]. Since there are no systematic trends in the size of the background, we do not consider it relevant to the superconducting DOS. As discussed in more detail in Ref. [10], we remove this background conductance in our data analysis by dividing the zero-field conductance by the high-field conductance, thus isolating the superconducting DOS. Using this procedure, we find that the resultant DOS satisfies the sum rule on the total density of states, except in our thickest samples, i.e. those with the smallest conductance variations.

The results of this normalization procedure for all measured d_F are shown in Fig. 1, with $d_F = 0$ (tunneling into pure Nb) at the bottom and $d_F = 5.0$ nm at the top, where the conductance scale has been magnified by roughly a factor of 1000 in comparison. The $d_F = 0$ curve has two clear BCS coherence peaks at $\pm\Delta_0 = 1.3$ meV; above that, we see that the addition of just 1.0 nm of Ni increases the ZBC significantly. In addition to the general trend of decreasing feature size as a function of d_F , for $d_F = 1.5$ nm we see a striking new feature: two coherence peaks on either side of zero-bias. Continuing up Fig. 1, we see this “double-peak” structure of the DOS for $d_F = 1.5 - 3.0$ nm, with the interior peaks moving to lower voltages as d_F increases. At $d_F = 3.5$ nm, though, we see a qualitatively different DOS: the coherence peaks at $V = \pm\Delta_0$ have “inverted” and are now conductance minima, while a narrow interior gap remains. This inverted DOS was seen in the first reported measurement of the DOS in S/F bilayers [14], yet has not been seen in other reports [10, 15]. The outer peak remains inverted at $d_F = 4.0$ nm, but by $d_F = 4.5, 5.0$ nm we have returned to the non-inverted DOS with the addition of a very narrow peak at zero-bias. These measurements of thicker samples ($d_F \geq 4.5$ nm) must be taken with a grain of salt as the feature size is quite small. It is our opinion that this zero-bias peak, which – like all other features of the DOS – does not change in a small parallel or perpendicular magnetic field is due to the steep voltage dependence of the background conductance and is therefore a by-product of our normalization procedure.

We model our system in the dirty limit, which is applicable only when the elastic scattering time, τ_e , is shorter than all other relevant time-scales. In the F-layer, $\hbar/\tau_e \approx 400$ meV (from resistivity data), which is much greater than either the estimated exchange field, $E_{ex} = 78$ meV (estimated from the Curie Temperature [16]), or the superconducting gap, $\Delta = 1.3$ meV. In this limit, the superconducting order parameter should

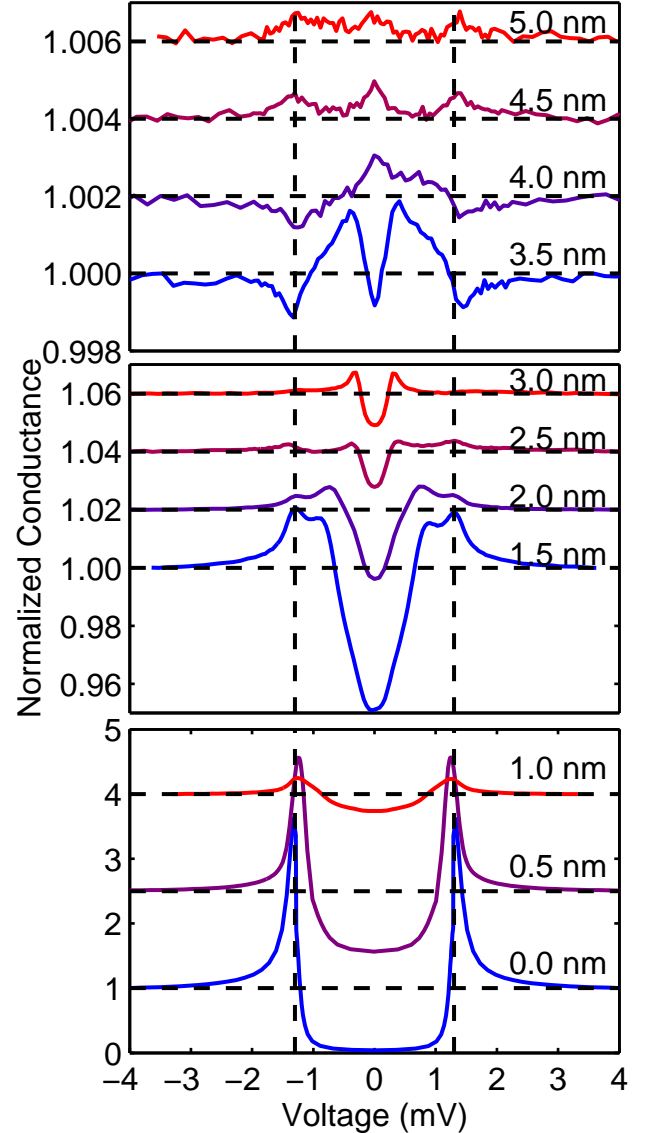


FIG. 1: Normalized conductances taken at 0.28 K for various Ni thickness indicated inside. From the bottom to the top plot the vertical scale is successively amplified. The curves are shifted for clarity. The vertical lines show that the outside peaks energy remains unchanged from 0.5 to 4.5 nm.

obey the Usadel equation [17, 18],

$$-\frac{\hbar D}{2} \frac{\partial^2 \theta_{\uparrow(\downarrow)}}{\partial x^2} + (-i\omega \pm iE_{ex} + 2\Gamma_Z \cos \theta_{\uparrow(\downarrow)}) \sin \theta_{\uparrow(\downarrow)} + \Gamma_X \sin(\theta_{\uparrow} + \theta_{\downarrow}) \pm \Gamma_{SO} \sin(\theta_{\uparrow} - \theta_{\downarrow}) = \Delta \cos \theta_{\uparrow(\downarrow)}, \quad (1)$$

where $\theta_{\uparrow(\downarrow)}$ corresponds to the up (down) electron density, D is the diffusion constant, ω is the energy, E_{ex} is the exchange field, Γ_Z and Γ_X are the strength of the magnetic scattering parallel and perpendicular to the quantization axis, and Γ_{SO} is the strength of the spin-orbit scattering. This equation is valid in the S-layer when all magnetic terms ($E_{ex}, \Gamma_Z, \Gamma_X$) are zero and in the F

layer when the gap, Δ , is zero. Δ must also obey the self-consistent equation, which at zero temperature is

$$\Delta(x) = \lambda \int_0^{\omega_D} d\omega (1/2) \text{Im} [\sin \theta_{\uparrow} + \sin \theta_{\downarrow}], \quad (2)$$

where ω_D is the Debye frequency, and λ is the coupling constant; this can be solved iteratively.

The S-layer and F-layer occupy the space $-d_S < x < 0$ and $0 < x < d_F$, respectively. We supplement our equation with the usual (non-magnetic) boundary conditions:

$$\left. \frac{d\theta_{\uparrow(\downarrow)}^F}{dx} \right|_{d_F} = 0, -\xi_N \gamma_B \left. \frac{d\theta_{\uparrow(\downarrow)}^F}{dx} \right|_{0+} = \sin(\theta_{\uparrow(\downarrow)}^S - \theta_{\uparrow(\downarrow)}^F) \quad (3)$$

$$\left. \frac{d\theta_{\uparrow(\downarrow)}^S}{dx} \right|_{-d_S} = 0, -\xi_S \frac{\gamma_B}{\gamma} \left. \frac{d\theta_{\uparrow(\downarrow)}^S}{dx} \right|_{0-} = \sin(\theta_{\uparrow(\downarrow)}^S - \theta_{\uparrow(\downarrow)}^F) \quad (4)$$

where $\xi_N = \sqrt{(\hbar D_F / 2\Delta)}$, $\xi_S = \sqrt{(\hbar D_S / 2\Delta)}$, $\gamma_B = R_B A / \rho_F \xi_N$, and $\gamma = \rho_S \xi_S / \rho_F \xi_N$. This notation is modeled after Refs. [18, 19], but with minor differences for ease of calculation. Finally, the total DOS measured by our junctions, $N(\omega)$, is

$$N(\omega) = (1/2) \text{Re} [\cos \theta_{\uparrow}^F(\omega) + \cos \theta_{\downarrow}^F(\omega)]_{x=d_F}. \quad (5)$$

If we numerically solve the Usadel equation with $E_{ex} \gg \Delta$ and no scattering, we find the characteristic decay and oscillation of the DOS as a function of d_F [20], but as d_F increases, it goes from *inverted* to *normal*, to *inverted*, etc. In other words, we expect the inverted — not the normal — DOS to be seen for thin ($0.2 \text{ nm} \lesssim d_F \lesssim 5 \text{ nm}$) F layers. Cottet *et al* [21] were the first to comment on this unusual prediction — which has not been seen in any tunneling study of S/F systems — and suggested that spin-dependent interfacial phase shifts (SDIPS) could resolve this disagreement.

We find that we can qualitatively account for the observed behavior through the addition of either Γ_Z , Γ_{SO} , or SDIPS — the quantitative fits discussed below favor Γ_{SO} as the dominant scattering term. Equally important, as noted before, we find that once the Usadel equation in the F-layer is essentially linear ($d_F \approx \xi_F$) the only effect of increasing d_F is to scale the DOS at the interface [10, 22]. This means that none of these parameters will produce any sub-gap structure — nor, for that matter, will any other parameter in Eq. 1 [25]

Thus, we are motivated to split empirically our DOS into two parts: a DOS with a single peak (or inverted peak) at $\pm\Delta_0$, which we call the outer gap, and whatever remains, which we call the sub gap. Since the $d_F = 1.0 \text{ nm}$ curve only has peaks at $\pm\Delta_0$ we can use it as a template to isolate the outer-gap features of the other curves. We want to break down the other curves into a sum of two curves: one that is a scaled

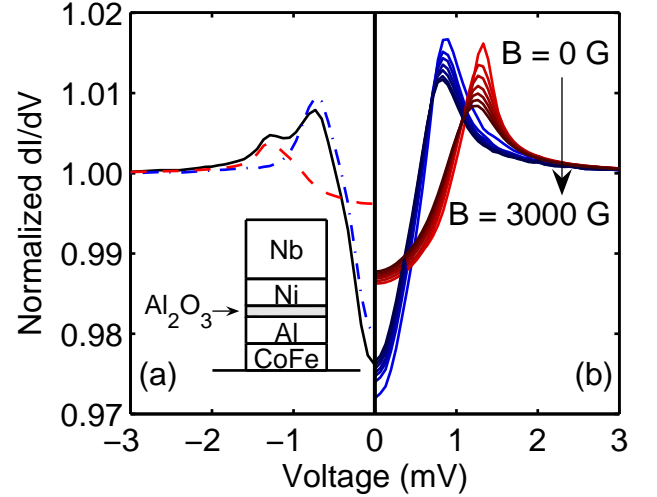


FIG. 2: (a) Normalized conductance of the $d_F = 2.0 \text{ nm}$ sample, split into *outer-gap* and *sub-gap* contributions. The outer-gap contribution (dashed line), a scaled version of the $d_F = 1.0 \text{ nm}$ curve and the sub-gap contribution (dash-dot line) sum to the total conductance (solid line). (b) Normalized conductance of the $d_F = 1.5 \text{ nm}$ sample taken in a perpendicular magnetic field, split into outer and sub-gap contributions. The sub-gap peak shifts in energy and changes amplitude slightly, but it does not broaden in an applied field, like the outer-gap.

(in the conductance-axis) copy of the template and another which will contain all of the sub-gap features. In order to determine the size of the different contributions, we adjust the scaling of the template such that the remaining sub-gap contribution is as smooth as possible at $\pm\Delta_0$. Figure 2 (a) shows the results of this process on the $d_F = 2.0 \text{ nm}$ sample. Qualitatively, the anomalous contribution for $1.5 \leq d_F \leq 3.5 \text{ nm}$ looks like a superconducting gap whose width decreases with increasing d_F and which stays un-inverted even when outer gap inverts.

Once we have isolated the outer-gap contribution for all d_F , we can analyze it quantitatively with the Usadel equation. As noted above, a combination of E_{ex} , SDIPS, Γ_Z , and Γ_{SO} could be used, but we find that the best and simplest fits require only E_{ex} and Γ_{SO} . Figure 3 shows the absolute value of $1 - N(0)$, the ZBC, versus d_F . The circles represent the ZBC of the outer-gap contribution and the triangles represent the sub-gap contribution. The line is the calculated ZBC from the Usadel equation with parameters $E_{ex} = 63 \text{ meV}$, $\Gamma_{SO} = 46 \text{ meV}$, $\gamma_B = 0.54$, and $\gamma = 0.52$, which fits the normal ZBC reasonably well [26]. Thus, we conclude that the outer gap is well-described by the Usadel equation with an exchange field and spin-orbit scattering.

We measured the $d_F = 1.5 \text{ nm}$ sample in a perpendicular magnetic field from $B = 0$ to 3000 G in 500 G increments. To isolate the outer gap, we measured the template curve ($d_F = 1.0 \text{ nm}$) at the same fields and used

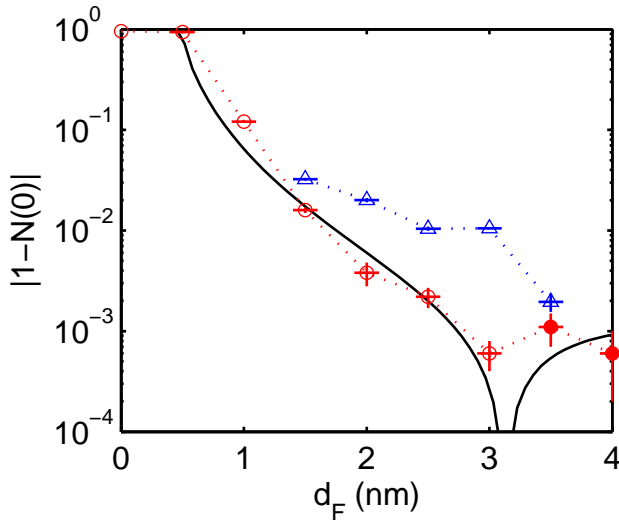


FIG. 3: $\text{Abs}(1 - N(0))$, the zero-bias conductance (ZBC), split into *outer-gap* (circles) and *sub-gap* (triangles) contributions. Open (filled) markers represent a ZBC less (greater) than unity. The outer-gap contribution is fitted to Eq. 1 with $E_{ex} = 63$ meV and $\Gamma_{SO} = 46$ meV (thin line).

those as templates for each applied field. Figure 2 (b) shows the results of this process: the outer curves are scaled versions of the template while the inner curves are the remaining sub-gap feature. The relative weight of the template needed to isolate each sub-gap curve is roughly constant, but the resulting sub-gap curves decrease in size with increasing field. Further, the shape of the sub-gap curve remains virtually unchanged in field, while the outer-gap peaks display significant broadening.

The existence of sub-gap structure alone indicates physics beyond the standard Usadel treatment. It is interesting to speculate what that might be. One intriguing possibility is that it is related to triplet superconductivity, perhaps in combination with some spin-flip scattering at the SF interface (see Bergeret *et al* [23] and references therein). While it is clearly premature to make this case confidently, we note that the relative insensitivity of the shape of the sub-gap peak to magnetic field and the apparent slower decay of its associated ZBC as a function of d_F are qualitatively consistent with such a possibility. Additional theory will be required to fully assess this possibility.

In summary, we have measured the DOS of Nb/Ni bilayers as a function of Ni thickness. In addition to tunneling features which are well-described by the Usadel equation with a strong exchange field and spin-orbit coupling, we have also discovered a robust sub-gap structure which cannot be explained by the conventional theory. By isolating this sub-gap contribution, we have shown that it behaves in a qualitatively different way from the Usadel contribution in a perpendicular magnetic field, which leads us to inquire whether it is the result of $m = 1$

triplet correlations in the bilayers.

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- [25] After discussion with A. Cottet, a newer model of SDIPS was created [24], which although promising does not produce any sub-gap structure for large d_S/ξ_S .
- [26] Applying a similar analysis to our previously published results on Nb/CoFe produces a reasonable fit for $E_{ex} = 109$ meV and $\Gamma_{SO} = 85$ meV. The large value of Γ_{SO} is necessary to account for the lack of an inversion.